



**PATENT APPLICATION**

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re application of

Docket No: Q66019

Kazuyuki SATO, et al.

Appln. No.: 09/914,835

Group Art Unit: 1712

Confirmation No.: 4395

Examiner: Margaret G. Moore

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For: SURFACE TREATMENT AGENT COMPRISING ORGANIC-INORGANIC HYBRID MATERIAL

**DECLARATION UNDER 37 C.F.R. § 1.132**

Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Sir:

I, Fumihiko Yamaguchi, hereby declare and state:

THAT I am a citizen of Japan;

THAT I was graduated from the Department of Chemical Engineering, the Faculty of Engineering, Tokyo Institute of Technology, Japan, in March 1981, and I was also graduated from graduate school of Tokyo Institute of Technology, and received a master's degree in Engineering in March 1983;

THAT Since April 1983 up to the present time, I have been employed by Daikin Industries Ltd. and engaged in research works on developments of a fluoropolymer and a composition containing the fluoropolymer, where I hold a position as a section chief researcher;

THAT I am a co-inventor of the present invention; and

THAT the following experiments were carried out under my direct supervision.

EXAMPLES

Synthesis Example 3

The same procedure as in Synthesis Example 2 was followed to give Product 5 except that 15 parts of titanium (IV) ethoxide  $[\text{Ti}(\text{OCH}_2\text{CH}_3)_4]$  was used instead of 15 parts of tetraethoxysilane  $[\text{Si}(\text{OCH}_2\text{CH}_3)_4]$  (TEOS).

Synthesis Example 4

The same procedure as in Synthesis Example 2 was followed to give Product 6 except that 15 parts of zirconium (IV) propoxide  $[\text{Zr}(\text{OC}_3\text{H}_7)_4]$  was used instead of 15 parts of tetraethoxysilane  $[\text{Si}(\text{OCH}_2\text{CH}_3)_4]$  (TEOS).

Synthesis Example 5

The same procedure as in Synthesis Example 2 was followed to give Product 7 except that 15 parts of methyltriethoxysilane  $[\text{CH}_3\text{Si}(\text{OCH}_2\text{CH}_3)_3]$  was used instead of 15 parts of tetraethoxysilane  $[\text{Si}(\text{OCH}_2\text{CH}_3)_4]$  (TEOS), and 15 parts of 3-methacryloxypropyl dimethylmethoxy silane  $[\text{CH}_2=\text{CH}(\text{CH}_3)-(\text{C}=\text{O})-\text{O}-(\text{CH}_2)_3\text{Si}(\text{CH}_3)_2(\text{OCH}_3)]$  was used instead of 15 parts of 3-methacryloxypropyl trimethoxy silane  $[\text{CH}_2=\text{CH}(\text{CH}_3)-(\text{C}=\text{O})-\text{O}-(\text{CH}_2)_3\text{Si}(\text{OCH}_3)_3]$  (TMSM).

Synthesis Example 6

The same procedure as in Synthesis Example 2 was followed to give Product 8 except that 1.5 parts of perfluorooctanoic acid  $[\text{F}(\text{CF}_2)_7\text{COOH}]$  was used instead of 1.5 parts of 2-(perfluorooctyl)ethanol  $[\text{F}(\text{CF}_2)_8\text{CH}_2\text{CH}_2\text{OH}]$ .

Synthesis Example 7

The same procedure as in Synthesis Example 2 was followed to give Product 9 except that 1.5 parts of perfluorooctane sulfonic acid  $[\text{F}(\text{CF}_2)_7\text{SO}_3\text{H}]$  was used instead of 1.5 parts of 2-(perfluorooctyl)ethanol  $[\text{F}(\text{CF}_2)_8\text{CH}_2\text{CH}_2\text{OH}]$ .

#### Synthesis Example 8

The same procedure as in Synthesis Example 2 was followed to give Product 10 except that 2.0 parts of potassium perfluorooctanoate  $[\text{F}(\text{CF}_2)_7\text{COOK}]$  was used instead of 1.5 parts of 2-(perfluorooctyl)ethanol  $[\text{F}(\text{CF}_2)_8\text{CH}_2\text{CH}_2\text{OH}]$ , and 15 parts of polyacrylic acid (PA) aqueous solution (solid content: 40% by weight) was used instead of 15 parts of polymethacrylic acid (PMA) aqueous solution (a stain blocking agent, FX-668F manufactured by 3M Company).

#### Synthesis Example 9

The same procedure as in Synthesis Example 2 was followed to give Product 11 except that 1.5 parts of 3-(perfluorooctyl)-1,2-epoxypropane  $[\text{F}(\text{CF}_2)_8\text{CH}_2\text{CH}(\text{O})\text{CH}_2]$  was used instead of 1.5 parts of 2-(perfluorooctyl)ethanol  $[\text{F}(\text{CF}_2)_8\text{CH}_2\text{CH}_2\text{OH}]$ , and 5 parts of ethyl methacrylate (EMA) was used instead of 5 parts of methyl methacrylate (MMA).

Table A'

Additional Synthesis Examples 3 to 9

	Component (A)	Component (B)	Component (C)	Component (D)	Component (E)
Claims	metal alkoxide	Rf-X, X: group having reactive group -COOH, -SO <sub>3</sub> H, COOM, -SO <sub>3</sub> M, -OH, -CH(O)CH <sub>2</sub> , -PO <sub>3</sub> H <sub>2</sub> , -NCO, -NHCO-B	Polymer having group reactive with substrate	Metal chelate compound	Polymer- izable monomer
Synthesis Example 1	TEOS [Si(OCH <sub>2</sub> CH <sub>3</sub> ) <sub>4</sub> ]	CF <sub>3</sub> (CF <sub>2</sub> ) <sub>7</sub> CH <sub>2</sub> CH <sub>2</sub> -Si(OCH <sub>2</sub> CH <sub>3</sub> ) <sub>3</sub> (outside claims)	Polymethacrylic acid (PMA)	CH <sub>2</sub> =CH(CH <sub>3</sub> )-C(=O)O- (CH <sub>2</sub> ) <sub>2</sub> Si(OCH <sub>3</sub> ) <sub>2</sub>	MMA
Synthesis Example 2	TEOS [Si(OCH <sub>2</sub> CH <sub>3</sub> ) <sub>4</sub> ]	CF <sub>3</sub> (CF <sub>2</sub> ) <sub>7</sub> CH <sub>2</sub> CH <sub>2</sub> -OH	Polymethacrylic acid (PMA)	CH <sub>2</sub> =CH(CH <sub>3</sub> )-C(=O)O- (CH <sub>2</sub> ) <sub>2</sub> Si(OCH <sub>3</sub> ) <sub>2</sub>	MMA
Synthesis Example 3	Titanium (IV) ethoxide [Ti(OCH <sub>2</sub> CH <sub>3</sub> ) <sub>4</sub> ]	CF <sub>3</sub> (CF <sub>2</sub> ) <sub>7</sub> CH <sub>2</sub> CH <sub>2</sub> -OH	Polymethacrylic acid (PMA)	CH <sub>2</sub> =CH(CH <sub>3</sub> )-C(=O)O- (CH <sub>2</sub> ) <sub>2</sub> Si(OCH <sub>3</sub> ) <sub>3</sub>	MMA
Synthesis Example 4	Zirconium(IV) propoxide [Zr(OCH <sub>2</sub> CH <sub>3</sub> ) <sub>4</sub> ]	CF <sub>3</sub> (CF <sub>2</sub> ) <sub>7</sub> CH <sub>2</sub> CH <sub>2</sub> -OH	Polymethacrylic acid (PMA)	CH <sub>2</sub> =CH(CH <sub>3</sub> )-C(=O)O- (CH <sub>2</sub> ) <sub>2</sub> Si(OCH <sub>3</sub> ) <sub>3</sub>	MMA
Synthesis Example 5	Methyltriethoxy silane [CH <sub>3</sub> Si(OCH <sub>2</sub> CH <sub>3</sub> ) <sub>3</sub> ]	CF <sub>3</sub> (CF <sub>2</sub> ) <sub>7</sub> CH <sub>2</sub> CH <sub>2</sub> -OH	Polymethacrylic acid (PMA)	CH <sub>2</sub> =CH(CH <sub>3</sub> )-C(=O)O- (CH <sub>2</sub> ) <sub>2</sub> Si(CH <sub>3</sub> ) <sub>2</sub> (OCH <sub>3</sub> )	MMA
Synthesis Example 6	TEOS [Si(OCH <sub>2</sub> CH <sub>3</sub> ) <sub>4</sub> ]	CF <sub>3</sub> (CF <sub>2</sub> ) <sub>8</sub> COOH	Polymethacrylic acid (PMA)	CH <sub>2</sub> =CH(CH <sub>3</sub> )-C(=O)O- (CH <sub>2</sub> ) <sub>2</sub> Si(OCH <sub>3</sub> ) <sub>2</sub>	MMA
Synthesis Example 7	TEOS [Si(OCH <sub>2</sub> CH <sub>3</sub> ) <sub>4</sub> ]	CF <sub>3</sub> (CF <sub>2</sub> ) <sub>7</sub> SO <sub>3</sub> H	Polymethacrylic acid (PMA)	CH <sub>2</sub> =CH(CH <sub>3</sub> )-C(=O)O- (CH <sub>2</sub> ) <sub>2</sub> Si(OCH <sub>3</sub> ) <sub>2</sub>	MMA
Synthesis Example 8	TEOS [Si(OCH <sub>2</sub> CH <sub>3</sub> ) <sub>4</sub> ]	CF <sub>3</sub> (CF <sub>2</sub> ) <sub>8</sub> COOK	Polyacrylic acid (PA)	CH <sub>2</sub> =CH(CH <sub>3</sub> )-C(=O)O- (CH <sub>2</sub> ) <sub>2</sub> Si(OCH <sub>3</sub> ) <sub>2</sub>	MMA
Synthesis Example 9	TEOS [Si(OCH <sub>2</sub> CH <sub>3</sub> ) <sub>4</sub> ]	CF <sub>3</sub> (CF <sub>2</sub> ) <sub>7</sub> CH <sub>2</sub> CH(O)CH <sub>2</sub>	Polymethacrylic acid (PMA)	CH <sub>2</sub> =CH(CH <sub>3</sub> )-C(=O)O- (CH <sub>2</sub> ) <sub>2</sub> Si(OCH <sub>3</sub> ) <sub>2</sub>	EMA

The followings are additional Preparative Examples:

#### Preparative Example 7

#### Preparation of Product 5/MMA cross-linking precursor (Solution I)

The same procedure as in Preparative Example 1 was followed to give Product 5/MMA cross-linking precursor (Solution I) except that Product 5 was used instead of Product 1 used in the Preparative Example 1.

#### Preparative Example 8

#### Preparation of Product 6/MMA cross-linking precursor (Solution J)

The same procedure as in Preparative Example 1 was followed to give Product 6/MMA cross-linking precursor (Solution J) except that Product 6 was used instead of Product 1 used in the Preparative Example 1.

**Preparative Example 9**

**Preparation of Product 7/MMA cross-linking precursor (Solution K)**

The same procedure as in Preparative Example 1 was followed to give Product 7/MMA cross-linking precursor (Solution K) except that Product 7 was used instead of Product 1 used in the Preparative Example 1.

**Preparative Example 10**

**Preparation of Product 8/MMA cross-linking precursor (Solution L)**

The same procedure as in Preparative Example 1 was followed to give Product 8/MMA cross-linking precursor (Solution L) except that Product 8 was used instead of Product 1 used in the Preparative Example 1.

**Preparative Example 11**

**Preparation of Product 9/MMA cross-linking precursor (Solution M)**

The same procedure as in Preparative Example 1 was followed to give Product 9/MMA cross-linking precursor (Solution M) except that Product 9 was used instead of Product 1 used in the Preparative Example 1.

**Preparative Example 12**

**Preparation of Product 10/MMA cross-linking precursor (Solution N)**

The same procedure as in Preparative Example 1 was followed to give Product 10/MMA cross-linking precursor (Solution N) except that Product 10 was used instead of Product 1 used in the Preparative Example 1.

### Preparative Example 13

#### Preparation of Product 11/MMA cross-linking precursor (Solution O)

The same procedure as in Preparative Example 1 was followed to give Product 11/MMA cross-linking precursor (Solution O) except that Product 11 was used instead of Product 1 used in the Preparative Example 1.

Table A'

	Preparative Example						Additional Preparative Example						
	1	2	3	4	5	6	7	8	9	10	11	12	13
DMP	-	-	-	-	30	30	-	-	-	-	-	-	-
Pure water	-	-	-	-	401.3	401.3	-	-	-	-	-	-	-
LSH	-	-	-	-	3	3	-	-	-	-	-	-	-
N-17	-	-	-	-	4.5	4.5	-	-	-	-	-	-	-
HS-220	-	-	-	-	6	6	-	-	-	-	-	-	-
LT-221	-	-	-	-	4.5	4.5	-	-	-	-	-	-	-
APS	-	-	0.75	0.75	0.75	0.75	-	-	-	-	-	-	-
Product 1	90	-	90	-	90	-	-	-	-	-	-	-	-
Product 2	-	90	-	90	-	90	-	-	-	-	-	-	-
Product 3	-	-	-	-	-	-	-	-	-	-	-	-	-
Product 4	-	-	-	-	-	-	-	-	-	-	-	-	-
Product 5	-	-	-	-	-	-	90	-	-	-	-	-	-
Product 6	-	-	-	-	-	-	-	90	-	-	-	-	-
Product 7	-	-	-	-	-	-	-	-	90	-	-	-	-
Product 8	-	-	-	-	-	-	-	-	-	90	-	-	-
Product 9	-	-	-	-	-	-	-	-	-	-	90	-	-
Product 10	-	-	-	-	-	-	-	-	-	-	-	90	-
Product 11	-	-	-	-	-	-	-	-	-	-	-	-	90
DMF	-	-	450	450	-	-	-	-	-	-	-	-	-
Methanol	450	450	-	-	-	-	450	450	450	450	450	450	450
Benzoin methyl Ether	0.75	0.75	-	-	-	-	0.75	0.75	0.75	0.75	0.75	0.75	0.75
Methylene bis acrylamide	4	4	-	-	-	-	4	4	4	4	4	4	4

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The followings are additional Examples:

**Example 7**

The Solution I prepared in Preparative Example 7 was diluted with methanol to give a treatment liquid having a solid content of 3 % by weight. This treatment liquid was evaluated as in Example 1.

The results are shown in Table C'.

**Example 8**

The Solution J prepared in Preparative Example 8 was diluted with methanol to give a treatment liquid having a solid content of 3 % by weight. This treatment liquid was evaluated as in Example 1.

The results are shown in Table C'.

**Example 9**

The Solution K prepared in Preparative Example 9 was diluted with methanol to give a treatment liquid having a solid content of 3 % by weight. This treatment liquid was evaluated as in Example 1.

The results are shown in Table C'.

**Example 10**

The Solution L prepared in Preparative Example 10 was diluted with methanol to give a treatment liquid having a solid content of 3 % by weight. This treatment liquid was evaluated as in Example 1.

The results are shown in Table C'.

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**Example 11**

The Solution M prepared in Preparative Example 11 was diluted with methanol to give a treatment liquid having a solid content of 3 % by weight. This treatment liquid was evaluated as in Example 1.

The results are shown in Table C'.

**Example 12**

The Solution N prepared in Preparative Example 12 was diluted with methanol to give a treatment liquid having a solid content of 3 % by weight. This treatment liquid was evaluated as in Example 1.

The results are shown in Table C'.

**Example 13**

The Solution O prepared in Preparative Example 13 was diluted with methanol to give a treatment liquid having a solid content of 3 % by weight. This treatment liquid was evaluated as in Example 1.

The results are shown in Table C'.



Table C'

		Example						Additional Example						
		1	2	3	4	5	6	7	8	9	10	11	12	13
Before-cleaning	Water repellency	50	50	50	50	50	50	50	50	50	50	50	50	50
	Oil repellency	3	3	3	3	3	3	3	3	3	3	3	3	3
	SB property	10	10	10	10	10	10	10	10	10	10	10	10	10
	Soil releasability (%)	80	80	77	77	77	77	77	77	74	80	80	80	77
After cleaning (five times cleaning)	Water repellency	50	50	50	50	50	50	50	50	50	50	50	50	50
	Oil repellency	3	3	3	3	3	3	3	3	3	3	3	3	3
	SB property	10	10	10	10	10	10	10	10	10	10	10	10	10
	Soil releasability (%)	77	77	74	74	74	74	74	74	71	77	77	74	74
	Fluorine residual ratio (%)	95	95	95	95	95	95	95	95	95	95	95	90	95
Knoop hardness		33	33	30	30	28	28	30	30	28	33	33	33	30

I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: December 14, 2004

  
Mr. Fumihiko Yamaguchi